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Nuclear Capture Reactions for Stockpile Stewardship and Basic Science

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Introduction

The capture process is a nuclear reaction in which a target atom captures an incident projectile, *e.g.* a neutron. The excited-state compound nucleus de-excites by emitting photons. This process creates an atom that has one more neutron than the target atom, so it is a different isotope of the same element. With low energy (slow) neutron projectiles, capture is the dominant reaction, other than elastic scattering. However, with very heavy nuclei, fission competes with capture as a method of de-excitation of the compound nucleus. With higher energy (faster) incident neutrons, additional reactions are also possible, such as emission of protons or emission of multiple neutrons. The probability of a particular reaction occurring (such as capture) is referred to as the cross section for that reaction. Cross sections are very dependent on the incoming neutron's energy.

Capture reactions can be studied either using monoenergetic neutron sources or "white" neutron sources. A "white" neutron source has a wide range of neutron energies in one neutron beam. The advantage to the white neutron source is that it allows the study of cross sections as they depend on neutron energies. The Los Alamos Neutron Science Center, located at Los Alamos National Laboratory, provides an intense white neutron source.[1] Neutrons there are created by a high-energy proton beam from a linear accelerator striking a heavy metal (tungsten) target. The neutrons range in energy from subthermal up to very fast - over 100 MeV in energy.

Low-energy neutron reaction cross sections fluctuate dramatically from one target to another, and they are very difficult to predict by theoretical modeling. The cross sections for particular capture reactions are important for defense sciences, advanced reactor concepts, transmutation of radioactive wastes and nuclear astrophysics. We now have a strong collaboration between Lawrence Livermore National Laboratory, Los Alamos National Laboratory, North Carolina State University and Charles University in Prague. In this paper, we report neutron capture studies that are of particular interest to Lawrence Livermore National Laboratory.

In addition to determining neutron capture cross sections, we are also interested in the nuclear properties of the excited state compound nuclei created in the capture reactions. One model that describes the behavior of the nucleus is the statistical model. Our statistical studies included measuring the photon strength function, resonance

parameters, level density and gamma-ray (γ-ray) cascade multiplicity. The DANCE array allows the separation of cascades by the number of transitions (multiplicity) in the cascade, and this makes it possible to study detailed properties of the statistical cascade such as the relationship between multiplicity and energy distributions.

The work reported here includes reaction on molybdenum targets, europium targets, gadolinium targets and the first americium-242m target. Our goal is to improve the accuracy and provide new measurements for stable and radioactive targets. We are especially interested in energy-dependent neutron capture cross sections.

In all of our experiments, the photons emitted in the capture reactions are gamma rays, and they are detected by the barium fluoride crystal array named the Detector for Advanced Neutron Capture Experiments (DANCE) shown in Fig. 1. The detector array is made of 160 crystals arranged in a sphere around the target. There are four different crystal shapes, each of which covers an equal solid angle. This array was specifically designed to measure neutron capture cross sections with targets that were milligram sized or smaller, including radioactive targets.[2] The barium fluoride crystals are scintillation (light generating) detectors with very fast response time, and are therefore suitable for high count rate experiments. Actual neutron capture events must be reliably distinguished from background γ-rays, which are always present in neutron induced reactions. To reduce the background of scattered neutrons, a lithium hydride shell is placed inside the array.

The purpose of using the spherical array of detectors is to cover all possible directions of emitted γ rays, so we will come as close as possible to complete detection of all the prompt γ-ray cascades emitted in a capture reaction. The sum of the energy of the γ cascades is a measure of the binding energy of the capture neutron. The binding energy is the energy required to remove a bound neutron from the nucleus. The measured mass of the nucleus is smaller than the masses of the target nucleus plus the captured neutron, and the difference (converted to energy) is the binding energy of the capture neutron. Because the detector is segmented into a large number of independent detectors, additional information on event multiplicities (number of γ rays emitted) and other properties can be determined.

Molybdenum experiments

The molybdenum-94 and molybdenum-95 capture reactions were studied for neutron energies from thermal to 16 keV. The primary motivation for these measurements was to test an enhancement observed in the low energy behavior of the photon strength function in molybdenum isotopes by the Oslo Cyclotron group.[3] The photon strength function is essentially a probability of nuclei to emit or absorb a γ-ray with a certain energy. The photon strength function is proportional to the photon transmission coefficient, which is an important quantity for accurate modeling of nuclear reaction cross sections, especially neutron capture cross sections.

Gamma-ray distribution and average γ cascade multiplicities were measured following neutron capture. These spectra were compared with computer simulations using the codes DICEBOX[4] and GEANT4. Models of the photon strength function with an Oslo type enhancement below 1 MeV were unable to reproduce experimental spectra. However, good agreement was found between DANCE spectra and simulations that postulate a low-lying resonance in the photon strength between the Kadmenskii-

Markushev-Furman (KMF) model and an Oslo-type photon strength function. The KMF model is a theoretical model which includes the temperature dependence of the Giant Electric Dipole Resonances (GEDR).

New spin assignments were made for resonances in the molybdenum-95 and molybdenum-96 excited nuclei. Assignments were made using the multiplicity of the γ-ray cascades and the γ-ray spectral shape. In the molybdenum compound nucleus, 56 new resonances were observed. Seventeen resonances were given a new spin assignment, and 22 resonances were given a tentative sin assignment. For the molybdenum-95 compound nucleus, 39 resonances were observed, and 21 resonances were given a new quantum number assignment (mainly parity assignment).[5] With the new resonance information, the resonance spacing can be improved. Accurate knowledge of resonance spacing is very important for reaction cross-section calculations because the resonance spacing is used to calculate the level density at the neutron binding energy; the accuracy of the level density determines the reliability of the calculated cross section. The new spacing is 20% lower than the previously reported spacing, which indicates a change in the cross section of as much as 40%.

Europium experiments

The europium-151 and europium-153 are naturally occurring stable europium isotopes. We selected these targets for study because there was a large 30-40% difference in existing data set.[6-8] The europium cross sections are very important for stockpile stewardship. The evaluated cross section data set used in modeling calculations is an average of the various disparate experimental measurements. New europium measurements were made using the DANCE array. New cross-section data, along with other measurements[9,10] are shown in Figs. 2 and 3. Results suggest that we should reduce the evaluated cross section for incident neutrons about 1 keV in energy.[11]

Various statistical properties of the europium-152 and europium-154 compound nuclei were investigated.[12] In the case of the europium targets, the multiplicity distributions (the probability distribution of the number of γ rays emitted following a neutron capture) were found to be independent of the incident neutron energy. The main discovery of the statistical study was the photon strength function. Simulations were performed, and the main finding was that, in the simulations, it was necessary to include an M1 scissors mode resonance (a mode in which the protons and neutrons collectively move against each other in a mode that resembles the movement of scissors). An independent experiment to confirm this finding had begun at the Charles University in Prague and the Nuclear Physics Institute (in Rez near Prague).

Gadolinium experiments

The gadolinium data is similar to the europium data in that there are several historical measurements which do not agree with each other. In particular, there are some differences within the gadolinium-154 neutron capture data. We have collected data on gadolinium-152 gadolinium-154, gadolinium -157 and natural gadolinium, the analysis of which is currently underway. We plan to measure gadolinium-160 in the summer of 2007. The gadolinium measurements will allow us to study several isotopes of the same element. Gadolinium and europium are both in the lanthanide series of the periodic table, a series which is very important to the astrophysical slow process.

Americium experiments

In our next experiment, we expect to measure the americium-242m capture cross section. For all the actinide targets, neutron bombardment results in both fission and capture reactions. In addition, the actinide elements (which include uranium, neptunium, plutonium, americium and several other heavier elements) undergo spontaneous decay by alpha emission. Gamma rays are emitted in both fission reactions and in alpha decay. Los Alamos National Laboratory, in collaboration with Lawrence Livermore National Laboratory, has developed a fission-tagging detector which tags, or marks, γ rays that were associated with fission nuclear reactions or alpha decay reactions.[13] Those γ rays may then be subtracted from the total γ spectrum, leaving only γ rays from capture reactions. Alternatively, the fission-tagging detector allows simultaneous measurement of fission and capture cross sections.

There are currently no capture cross-section data for americium-242m, which is an unusual isotope because it has both an odd number of protons and an odd number of neutrons in the nucleus. It is also the only odd-odd actinide for which we have target material. The americium-242m target material was made in the Idaho National Laboratory's reactor by neutron bombardment of americium-241. A reactor neutron plus americium-241 produces americium-242 plus the metastable americium-242m. The americium-242 decays with a half-life of 16 hours, leaving the americium-242m, which has a half-life of 141 years. Our supply of target material is unique in the world. Samples were recently purified and electroplated onto thin beryllium foils.

We made a preliminary measurement on a 47 μg target in September of 2006. With this thin target, we were able to make a good Am-242m fission cross section measurement. After subtraction of the γ rays associated with fission, we found that the capture data was barely above the background. Above 7 eV, the DANCE experimental data is in excellent agreement with the previous neutron induced fission cross-section measurements done by J. Browne, *et al.*[14] Discrepancies between the DANCE data and the Browne data are currently being studied.

During 2007, we plan to make a new thicker americium target by electroplating material onto titanium foils, and we expect that the total target material will be 400 μg of americium.

Summary

DANCE is an excellent facility for determining capture cross sections and for studying statistical decay of excited nuclei. Our molybdenum experiment was a good test case for improving the spin assignments, and we were able to contribute to the discussion regarding the molybdenum strength function. We were able to determine europium-151 and europium-153 neutron capture cross sections over a wide neutron energy range, and we used our data for statistical decay studies. Neutron capture data has also been collected for several gadolinium isotopes. With the fission-tagging detector, we made a new measurement of the americium-242m fission cross section. The americium-242m neutron capture cross-section measurement will be the first of an odd-odd actinide.

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Figures

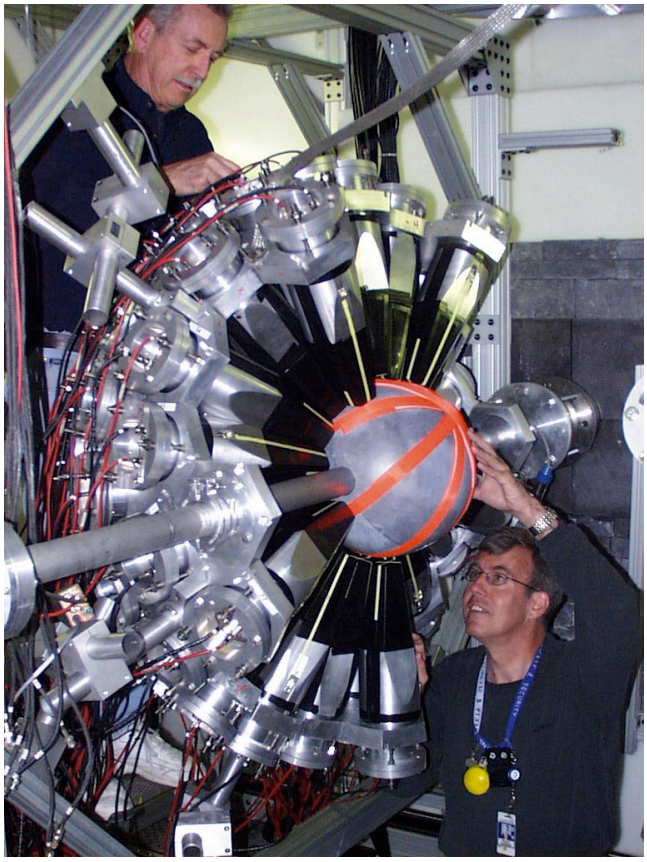

Fig. 1. The DANCE detector array is made of 160 barium fluoride crystals. The array can be pulled apart; half of the crystals are shown here

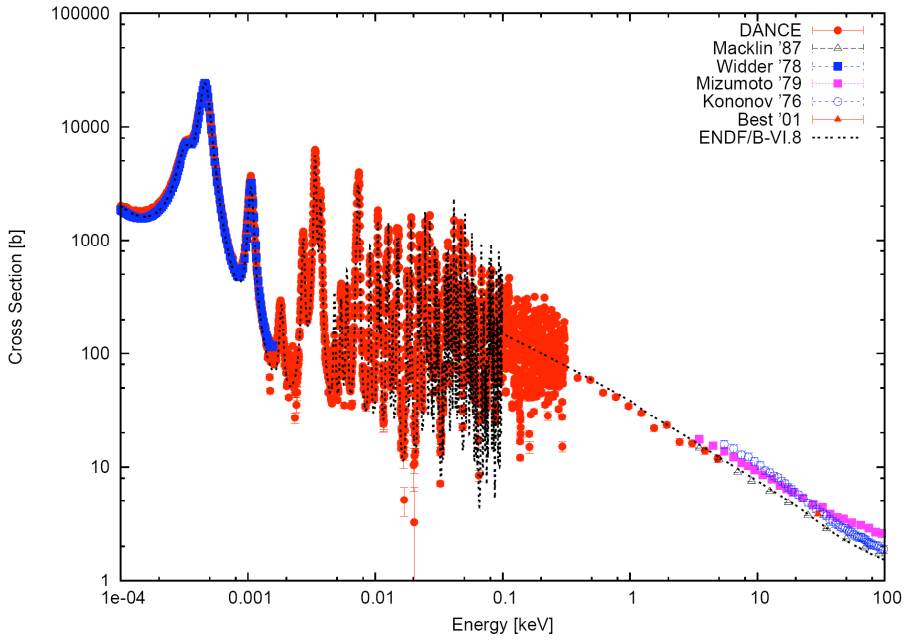

Fig. 2. The new europium-151 data set with several other europium-151 capture gamma measurements. The DANCE data is shown in red circles. The new data agrees with the 1987 Macklin data.

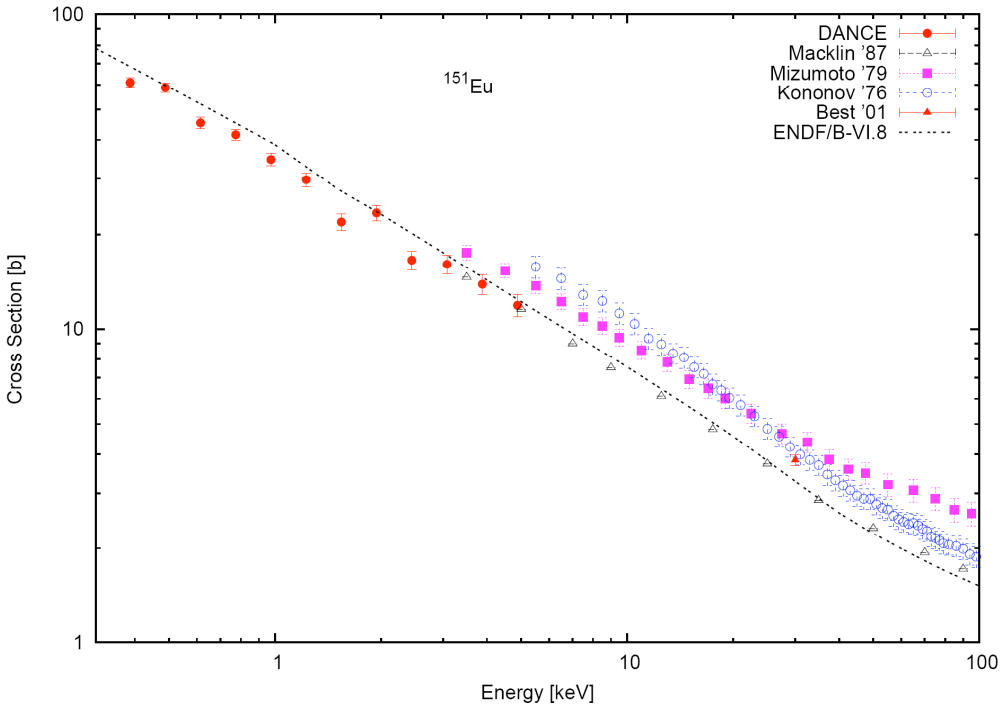

Fig. 3. Neutron capture cross-section data for europium-151 from 0.3 keV to 100 keV. The ENDF/B-VI calculation agrees with the new DANCE data and the Macklin data.